Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

OVERALL SEPARATION FACTORS FOR STABLE ISOTOPES BY GAS CENTRIFUGE

Chuntong Ying[a]; Shi Zeng[a]; Yuguang Nie[a]; Xiuyong Shang[a]; Houston G. Wood[b]
[a] Department of Engineering Physics, Tsinghua University, Beijing, People's Republic of China [b] Department of Mechanical & Aerospace Engineering, University of Virginia, Charlottesville, Virginia, U.S.A.

Online publication date: 03 December 2001

To cite this Article Ying, Chuntong , Zeng, Shi , Nie, Yuguang , Shang, Xiuyong and Wood, Houston G.(2001) 'OVERALL SEPARATION FACTORS FOR STABLE ISOTOPES BY GAS CENTRIFUGE', Separation Science and Technology, 36: 2, 159 — 175

To link to this Article: DOI: 10.1081/SS-100001073

URL: http://dx.doi.org/10.1081/SS-100001073

SEPARATION SCIENCE AND TECHNOLOGY, 36(2), 159–175 (2001)

OVERALL SEPARATION FACTORS FOR STABLE ISOTOPES BY GAS CENTRIFUGE

Chuntong Ying,[1,*] Shi Zeng,[1] Yuguang Nie,[1] Xiuyong Shang,[1] and Houston G. Wood[2]

[1]Department of Engineering Physics, Tsinghua University, Beijing, People's Republic of China
[2]Department of Mechanical & Aerospace Engineering, University of Virginia, Charlottesville, Virginia

ABSTRACT

The demand for stable isotopes is stimulating theoretical and experimental research on separation of stable isotopes by gas centrifuge. Most of the stable elements in nature have three or more isotopes. For modern gas centrifuges the separation factors usually are not as close to unity as is the case for the gaseous diffusion process. The overall separation factor for the unit molar weight difference, γ_0, is an important parameter for describing the separation performance of a gas centrifuge, and it depends on many variables of the gas centrifuge and physical properties of process gases. In this paper examples of the influence of some parameters on the overall separation factor will be shown.

Key Words: Overall separation factor; Gas centrifuge

*Corresponding author. E-mail: yingct@mail.tsinghua.edu.cn

INTRODUCTION

Recently, the demand has been growing for stable isotopes in physical and chemical research and in medical diagnostics. The gas centrifuge process has made it possible to produce many nonuranium isotopes economically, especially when large quantities are needed. Many countries, such as the United States (1,2), Russia (3–5), and China (6), and organizations, such as URENCO (7), have reported their activities in the public literature in the field of multicomponent separation by gas centrifuge.

Most of the 84 stable elements that exist in the nature have three or more isotopes. For modern gas centrifuges, the separation factors usually are not as close to unity as is the case for gaseous diffusion process. The separation factors of a gas centrifuge between the *i*th and the *j*th isotopes, γ_{ij}, may be expressed as $\gamma_{ij} = \gamma_0^{M_j - M_i}$, where γ_0 is the overall separation factor for the unit molar weight mass difference. The overall separation factor, γ_0, is an important factor to describe the separation characteristic of a gas centrifuge, and γ_0 depends on a lot of variables, such as the angular velocity of the cylinder, Ω, the length of the cylinder, Z_H, the feed of the gas centrifuge, F, etc. For different process gas γ_0 depends on its physical properties, such as ρD, Where ρ is density of working media and D is its diffusion coefficient, the viscosity of process gas, μ, etc.

Because the overall separation factor γ_0 is an important separation characteristic parameter, it is of interest to know among the many variables which are the key ones. In this paper, examples of the influence of some parameters on the overall separation factor are shown. The main variables for the overall separation factor for a given gas centrifuge are: feed flow rate to the gas centrifuge, the product ρD, and the parameter A^2.

THEORETICAL ANALYSIS

A schematic of a gas centrifuge is shown in Figure 1. For the separation factors we use the following definitions:

$$\alpha_{ij} \equiv \frac{C_i^P}{C_i^F} \Big/ \frac{C_j^P}{C_j^F}; \quad \beta_{ij} \equiv \frac{C_i^F}{C_i^W} \Big/ \frac{C_j^F}{C_j^W}; \quad \gamma_{ij} = \alpha_{ij} * \beta_{ij} = \frac{C_i^P}{C_i^W} \Big/ \frac{C_j^P}{C_j^W} \qquad (1)$$

The separation factor for a mixture of n isotopes, γ_{ij}, has the following relationship with the molar weight of the components (13):

$$\gamma_{ij} = \gamma_0^{M_j - M_i} \quad i = 1, \ldots n; \quad j = 1, \ldots n \qquad (2)$$

where M_i is the molar weight of the *i*th component, M_j is the molar weight of the

MARCEL DEKKER, INC.
270 Madison Avenue, New York, New York 10016

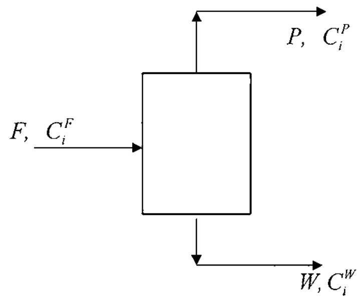

Figure 1. A schematic of a gas centrifuge.

*j*th component, γ_0 is the overall (heads to tails) separation factor per unit molar weight difference.

γ_0 depends on many parameters, such as the following values of a given gas centrifuge:

The length of the gas centrifuge Z_H
The radius of the gas centrifuge r_a
The peripheral rotation speed Ωr_a
The axial location of the feed point Z_F

Besides these parameters, other parameters or factors that have influence on the overall separation factor, γ_0, are:

The feed rate of the process gas F
The cut θ
The temperature T_0 and the temperature distribution of the temperature on the wall
The parameters of the scoops
The pressure of the process gas at the rotor wall p_W
The viscosity of the process gas μ
The molar weight of the process gas M
The product of ρD, where ρ is density of process gas and D is its diffusion coefficient

Examples have been considered for four different isotopic mixtures: UF_6 for uranium separation, WF_6 for tungsten separation, OsO_4 for osmium separation, and Xe for xenon separation. A number of authors have discussed the separation of multicomponent isotopes by gas centrifuge (8–14). The method described by Ying et al. (13) has been used in the present work to calculate the four examples of separation in a given gas centrifuge. The governing composition equations in a single gas centrifuge is (13)

$$\left(\frac{1}{2\pi\rho\mathcal{D}_i}\int_0^{r_a}\frac{\psi^2}{r}\,dr + \pi\rho\mathcal{D}_i r_a^2\right)\frac{d\bar{C}_i}{dz} = \frac{\Omega^2}{RT}(\bar{M}-M_i)C_i\int_0^{r_a}\psi r\,dr - \left(\frac{\bar{M}}{M_i}P_i^* - P^*\bar{C}_i\right) \quad i = 1, 2, \ldots n \quad (3)$$

where $\bar{C}_i$ is the radial averaged composition of the ith isotope in a multicomponent mixture with n components, P^* is the net axial flow flux of the mixture, P_i^* is the net axial flow flux of the ith component, ψ is the stream function $\psi \equiv \int_0^r \rho V_z 2\pi r\,dr$, V_z is the axial component of the velocity of the mixture in the gas centrifuge, $\bar{M}$ is the average molecular weight of the mixture, that is $\bar{M} = \sum_{i=1}^n M_i C_i$, $\mathcal{D}_i$ is defined as (13)

$$\mathcal{D}_i = \left(\sum_{k=1}^n \frac{C_k}{\mathcal{D}_{ik}}\right)^{-1}$$

here $\mathcal{D}_{ik}$ is the binary diffusion coefficient.

With the following definitions:

$$\begin{aligned}
\varepsilon_i &\equiv \frac{\Omega^2 r_a^2}{2RT}(\bar{M}-M_i)\\
\varphi_{Pi} &\equiv \frac{\theta F}{\pi r_a \rho \mathcal{D}_i}; \quad \varphi_{Wi} \equiv \frac{(1-\theta)F}{\pi r_a \rho \mathcal{D}_i}\\
Y_{1i} &\equiv \frac{1}{r_a^2 \pi r_a \rho \mathcal{D}_i}\int_0^{r_a}\psi r\,dr \qquad (4)\\
Y_{2i} &\equiv \frac{1}{2(\pi r_a \rho \mathcal{D}_i)^2}\int_0^{r_a}\frac{\psi^2}{r}\,dr
\end{aligned}$$

Using the above-defined parameters, the composition equations in the enriching section of the gas centrifuge are obtained:

$$\begin{aligned}
(1+Y_{2i})\frac{dC_i}{ds} &= (2\varepsilon_i Y_{1i} + \varphi_{Pi})C_i - \frac{\bar{M}}{M_i}C_i^P\varphi_{Pi}; \quad I = 1, 2, \ldots n-1\\
C_n &= 1 - \sum_{i=1}^{n-1} C_i \qquad (5)
\end{aligned}$$

where $s = Z/r_a$, C_i^P is the composition of the ith component at the product end. The overbar of C is dropped.

The composition equations in the stripping section of the gas centrifuge are

$$(1 + Y_{2i})\frac{dC_i}{ds} = (2\varepsilon_i Y_{1i} - \varphi_{Wi})C_i + \frac{\bar{M}}{M_i}C_i^W \varphi_{Wi}; \quad I = 1, 2, \ldots n-1$$

$$C_n = 1 - \sum_{i=1}^{n-1} C_i \tag{6}$$

where C_i^W is the composition of the ith component at the waste end.

To solve the above equations, an iterative procedure is used (13). Before we start to solve Equations (5) and (6), we need to know the velocity distribution in the gas centrifuge, that is, the V_z or ψ. The velocity field is simulated by using Onsager's equation (15) solved by a finite method (16,17,18). As the paper (12) shows, the following partial differential equation is used:

$$(e^x(e^x \chi_{xx})_{xx})_{xx} + B^2 \chi_{yy} = F(x, y) \tag{7}$$

where χ is a master potential from which the physical variables, such as V_z, can be extracted, $x = A^2\ (1 - (r/r_a)^2)$ is the radial scale height or e-folding distance for the density, A^2 is a speed parameter ($A^2 \equiv M\Omega^2 r_a^2/2RT$), and y is the axial variable. The variable $B^2 \equiv \mathrm{Re}^2\ S/16A^{12}$ is a parameter containing the physical description of the particular cylinder and operation parameters. In particular, $\mathrm{Re} = \rho_W \Omega r_a^2/\mu$ where ρ_W is the density at the wall. The quantity $S = 1 + \mathrm{Pr}\ \ A^2(\gamma - 1)/2\gamma$ is a thermodynamic variable where γ is the ratio of specific heats and Pr is the Prandtl number. The nonhomogeneous term $F(x, y)$ arises from internal sources or sinks of mass, momentum, or energy and is written

$$\begin{aligned} F(x, y) = {} & \frac{B^2 A^2}{2ReS}\int_x^{x_T}(Z_y - 2V_y)\,dx' \\ & - \frac{B^2}{2ReS}\int_x^{x_T}\int_0^{x'}(Z_y + 2(S-1)V_y\,dx''\,dx' \\ & - \frac{B^2}{4A^4}\int_x^{x_T}\int_0^{x'} M_y\,dx''\,dx' - \frac{B^2A^2}{2ReS}[(e^x U_y)_x + (e^x W)_{xx}] \end{aligned} \tag{8}$$

Here *M, U, V, W*, and *Z* are dimensionless quantities that represent source terms in the modified forms of the conservation equations for mass, momentum, and energy. In terms of the dimensional physical variables, the source of mass is M, the source of momentum is $\mathbf{F}_s = (F_r, \mathrm{F}_\theta, \mathrm{F}_z)$, and sources of heat and work are Q and W. The mass introduced by the source has temperature T_s, velocity $\mathbf{V}_s = (V_r, \mathrm{V}_\theta, \mathrm{V}_z)$. The quantities in Equation (8) are related to these physical variables as follows:

$$
\begin{aligned}
M &= M/\rho_W\Omega \\
U &= (M v_r + F_r)/\rho_W\Omega^2 r_a \\
V &= (Re/4A^4)[(v_\theta - \Omega r)M + F_\theta]/\rho_W\Omega^2 r_a \\
W &= 2A^2[M v_z + F_z]/\rho_W\Omega^2 r_a \\
Z &= \frac{1}{4A^4}\left\{Q + W - \mathbf{q}\cdot\mathbf{F}_s + M\left[\frac{(V_s - q)^2}{2} - c_P(T_0 - T_s)\right]\right\}\Big/\left(kT_0 r_a^2\right)
\end{aligned}
\tag{9}
$$

Here **q** is the local velocity of the rotating gas, which is assumed to be given by solid body rotation, or $\mathbf{q} = (0, \Omega, 0)$.

Three drives of the countercurrent flow were considered. They are: scoop drive, wall temperature drive, and end cap drive. For this work, the code used for calculating the flow field is the one developed by Wei (16).

The results of calculation are shown in Figures 2 through 17. For all four examples, the composition of the feed flow is supposed to have the natural value, and the following parameters are kept fixed at the same value:

The length of the cylinder of the given gas centrifuge, Z_H
The radius of the cylinder, r_a
The angular velocity, Ω
The temperature T_0
The cut, θ, which equals 0.45

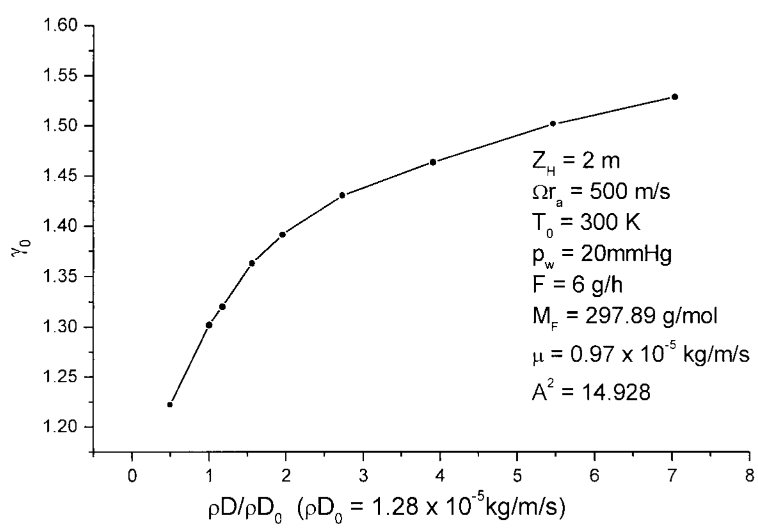

Figure 2. $\gamma_0 \sim \rho D$ for WF_6.

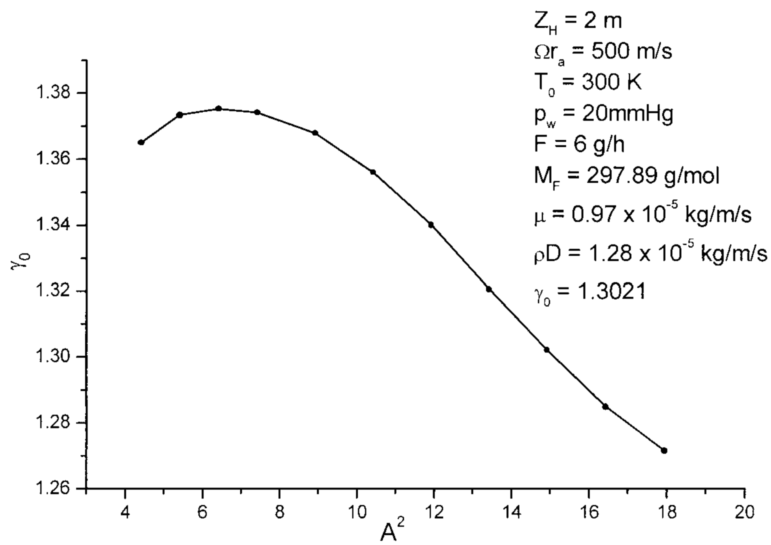

Figure 3. $\gamma_0 \sim A^2$ for WF_6.

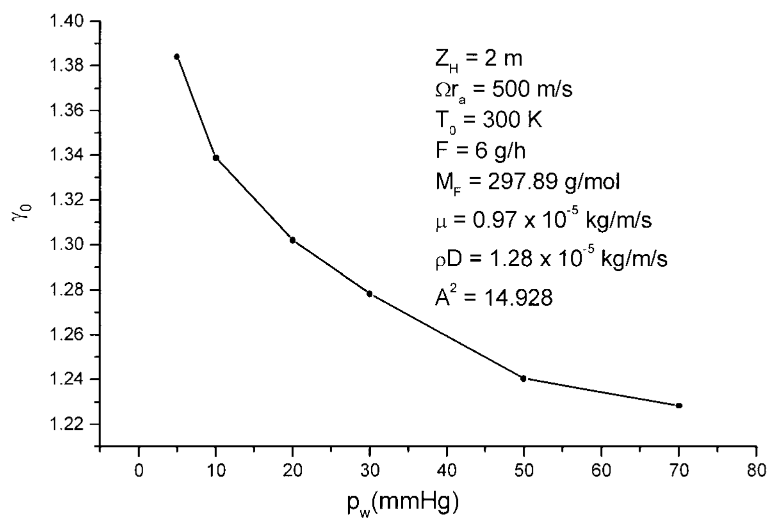

Figure 4. $\gamma_0 \sim p_w$ for WF_6.

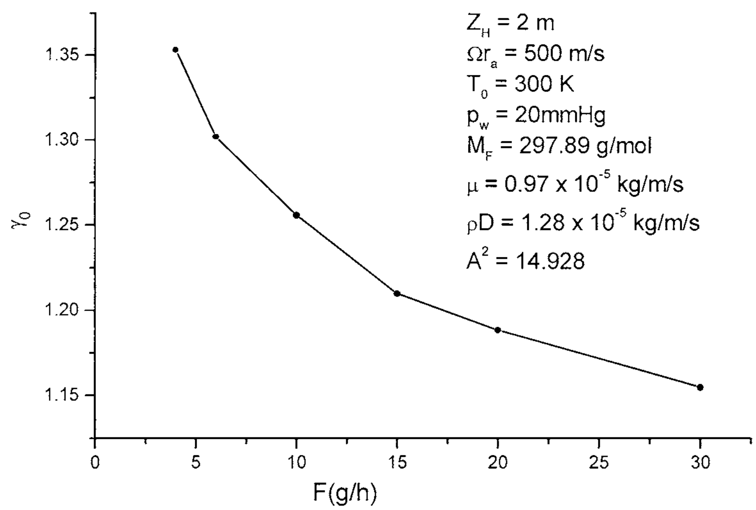

Figure 5. $\gamma_0 \sim F$ for WF_6.

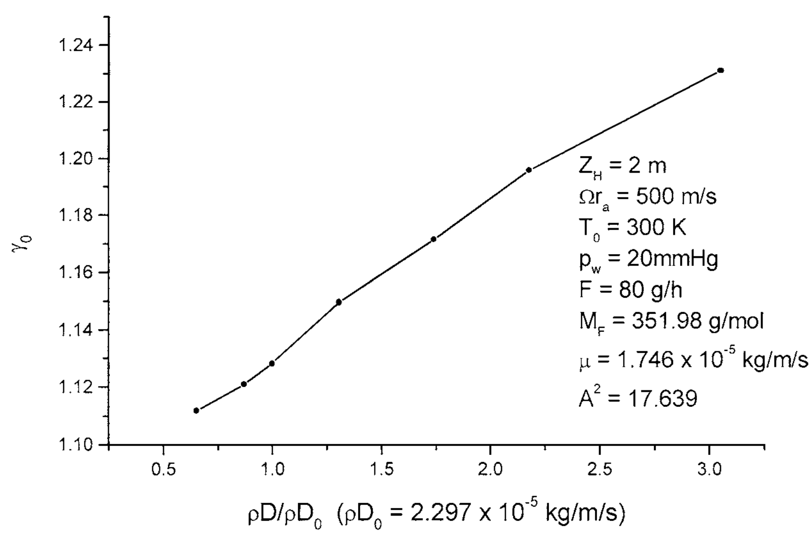

Figure 6. $\gamma_0 \sim \rho D$ for UF_6.

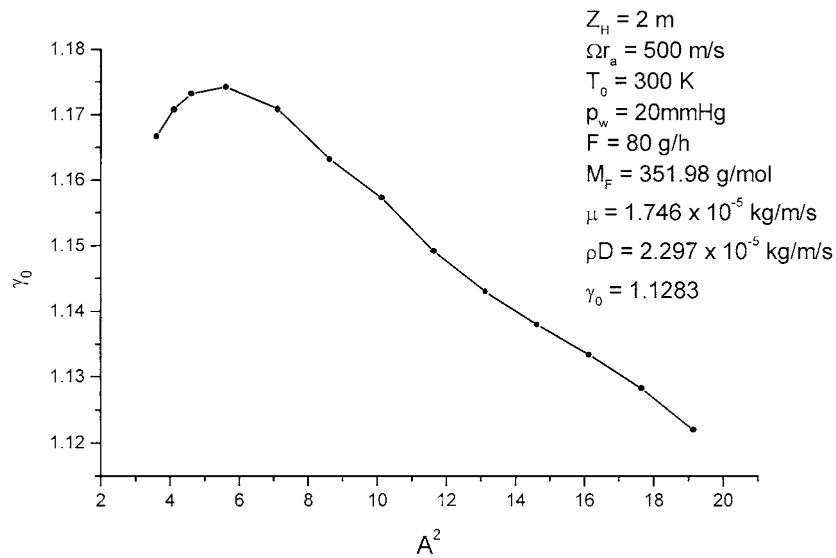

Figure 7. $\gamma_0 \sim A^2$ for UF_6.

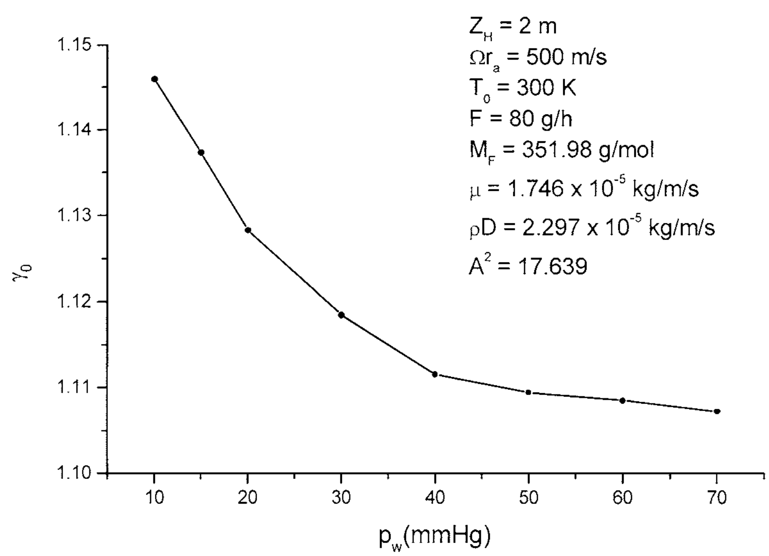

Figure 8. $\gamma_0 \sim p_w$ for UF_6.

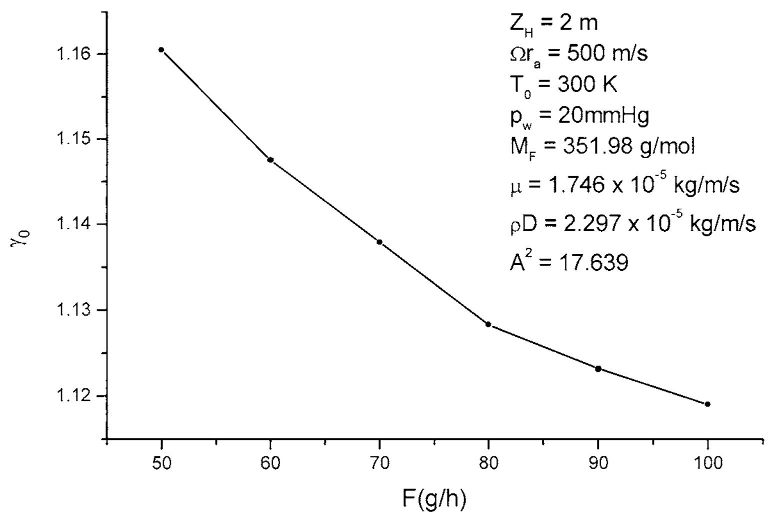

Figure 9. $\gamma_0 \sim F$ for UF_6.

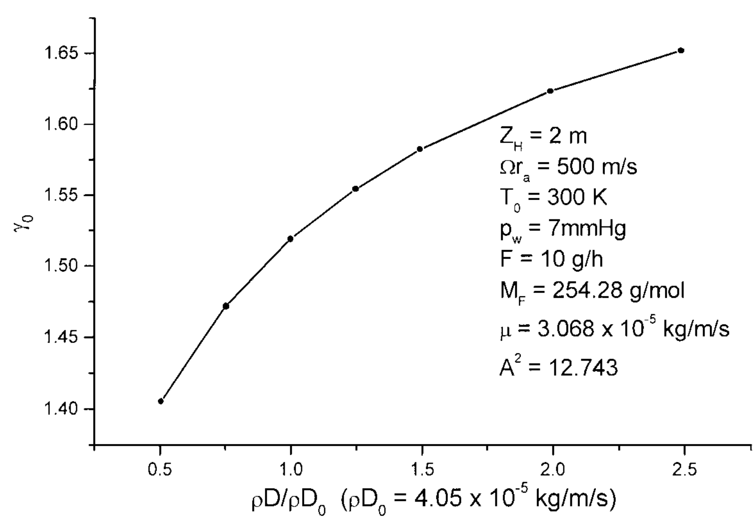

Figure 10. $\gamma_0 \sim \rho D$ for OsO_4.

Marcel Dekker, Inc.
270 Madison Avenue, New York, New York 10016

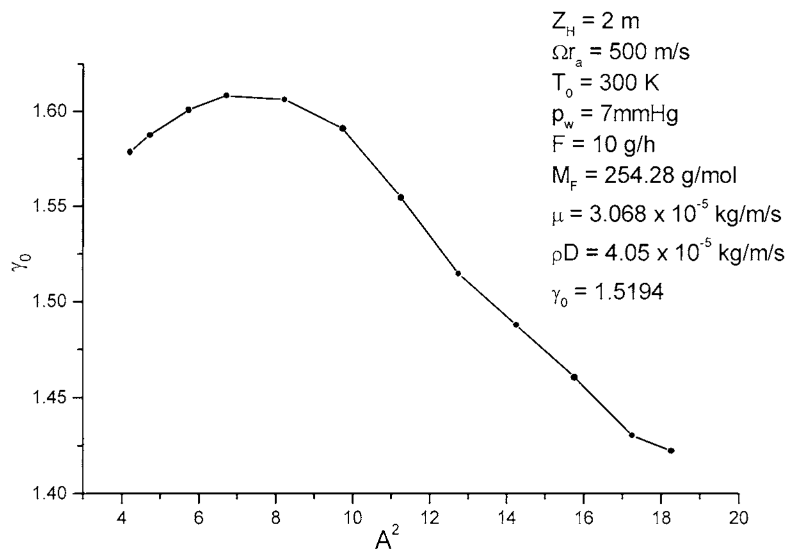

Figure 11. $\gamma_0 \sim A^2$ for OsO_4.

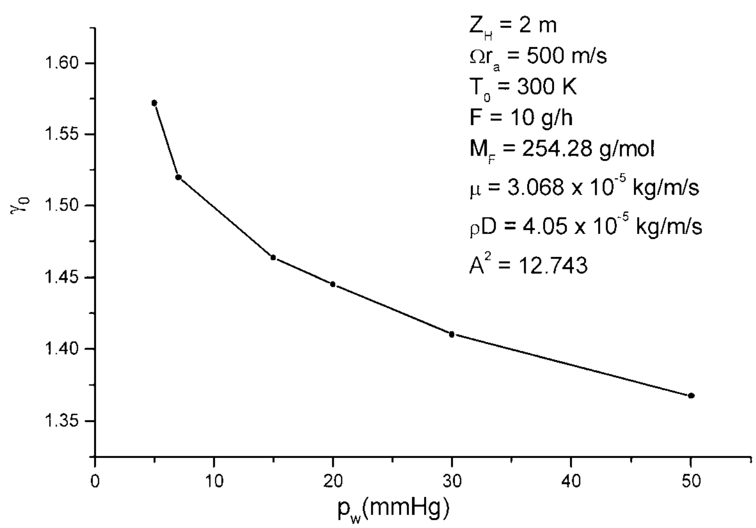

Figure 12. $\gamma_0 \sim p_w$ for OsO_4.

MARCEL DEKKER, INC.
270 Madison Avenue, New York, New York 10016

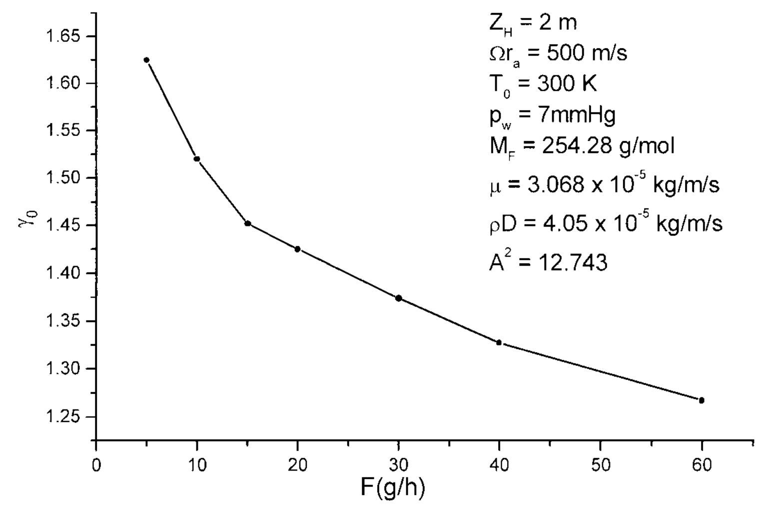

Figure 13. $\gamma_0 \sim F$ for OsO_4.

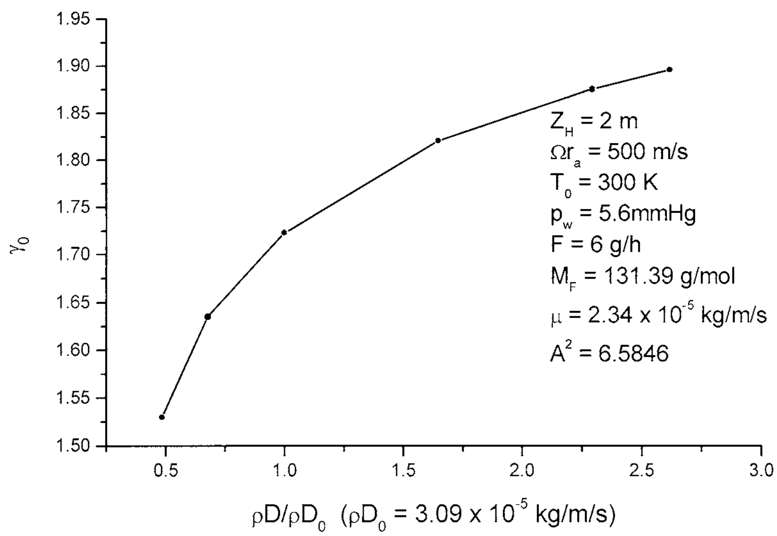

Figure 14. $\gamma_0 \sim \rho D$ for Xe.

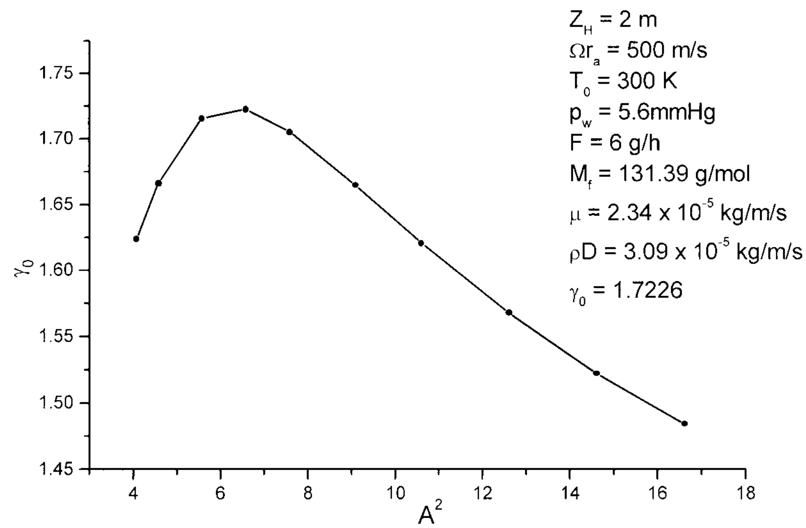

Figure 15. $\gamma_0 \sim A^2$ for Xe.

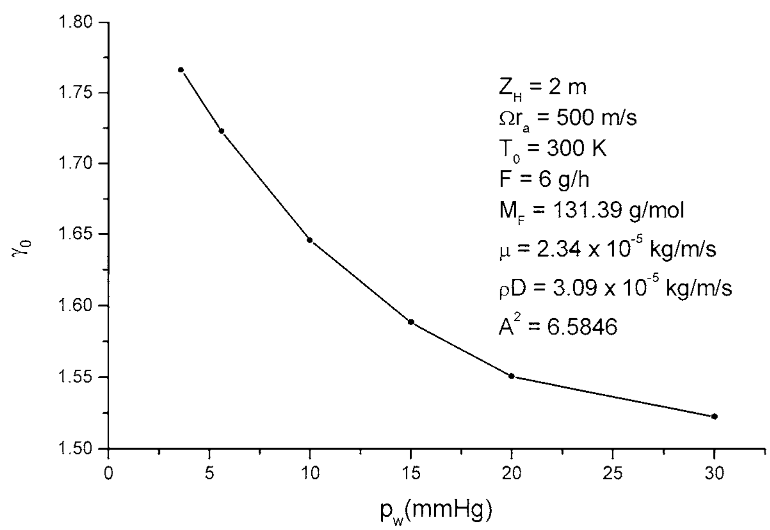

Figure 16. $\gamma_0 \sim p_w$ for Xe.

Marcel Dekker, Inc.
270 Madison Avenue, New York, New York 10016

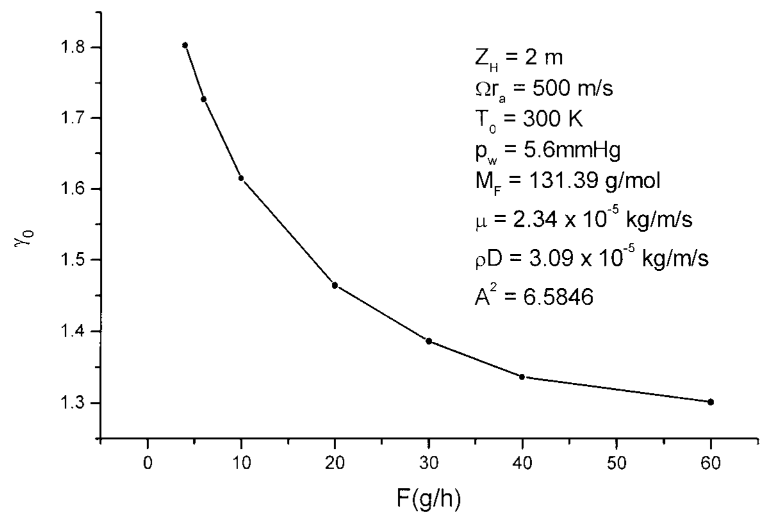

Figure 17. $\gamma_0 \sim F$ for Xe.

The values of the parameters are shown in the figures. In all figures M_F is the average molecular weight of the feed flow to gas centrifuge.

The curves in Figures 2 through 5 show the dependence of overall separation factor, γ_0, on different parameters, such as product ρD, the speed parameter A^2, pressure at wall p_W, and feed flow rate F, etc. The process gas is WF_6. For each curve, for instance the curve in Figure 2, which shows the dependence of γ_0 on ρD, the value of γ_0 is obtained by optimization of the three drives under the condition that all other parameters are given. The curve $\gamma_0 \sim \rho D$ is obtained from different given values of ρD. When we compute the value of γ_0 we use Equations (5), (6), and (3), where $\mathcal{D}_i$ is defined as an inverse of the summation of the ratio of mole fractions to binary diffusion coefficients. For simplification to express in figures, the D is a binary diffusion coefficient, corresponding the average molecular weight. In Figure 2 the value of ρD is scaled by ρD_0, which is the actual value of the process gas WF_6, and the value of ρD is a suppositional one. All the other curves are obtained in similar way. In Figure 3 the curve reaches its maximum value when A^2 is about 6. When the speed parameter, A^2, is not much greater than unity, the effect of the curvature must be considered and the partial differential equation of master potential is changed (17). All the consideration is included in the code we used. Figure 4 shows the $\gamma_0 \sim p_W$ curve, and Figure 5 shows the $\gamma_0 \sim F$ curve.

The results of UF_6 are shown in Figures 6 through 9. The results of OsO_4 are shown in Figures 10 through 13. The results of Xe are shown in Figures 14 through 17.

MARCEL DEKKER, INC.
270 Madison Avenue, New York, New York 10016

DISCUSSION AND CONCLUSIONS

The overall separation factor for unit molar weight difference, γ_0, is an important characteristic parameter for separation of multicomponent isotopes. Besides the construction parameters of the gas centrifuge, γ_0 depends on many variables. Some of them are the operating conditions, such as feed flow rate F, pressure at wall p_W. etc. Also, γ_0 depends on physical properties, such as molar weight M ($A^2 \equiv M\Omega^2 r_a^2/2RT$) and product ρD. From the examples, the parameters A^2 and ρD have great influence on the value of γ_0, and the operating condition feed flow rate has an effect on γ_0. From the figures it is obvious that the speed parameter A^2 plays a very important role for the value of γ_0, and the calculations show that γ_0 reaches its maximum when A^2 is about 6 for all the examples. We should say when we changed the value of A^2 only the M was changed; all the other parameters were kept fixed. The results presented in the figures show that γ_0 increases or decreases monotonically except for the $\gamma_0 \sim A^2$ curves.

The reason why A^2 and ρD have great influence might be explained if we use the idea of separative power, δU, for binary case. The separative power is a product of theoretical maximal separative power (19), δU_{max}, and several efficiency factors (20). The δU_{max} is proportional to ρD. One of the efficiency factors is flow pattern efficiency factor, which has a maximum near $A^2 = 6$. The explanation in detail should be an objective of another article.

ACKNOWLEDGMENT

Financial support was provided by the National Natural Science Foundation of People's Republic of China (No. 59676020).

REFERENCES

1. Roberts, W.L. Gas Centrifugation of Research Isotopes. Nucl. Instrum. Methods Phys. Res. **1989**, *A282*, 271–276.
2. Szady, A.J. Enrichment of Chromium Isotopes by Gas Centrifugation. Nucl. Instrum. Methods Phys. **1989**, *A282*, 277–280.
3. Borisevich, V.D.; Potapov, G.A.; Sulaberidze, G.A.; Chuzhinov, V.A. Multicomponent Isotope Separation in Cascade with additional External Flows. In *Proceedings of the Fourth Workshop on Separation Phenomena in Liquids and Gases*; Ying, C., Ed.; Tsinghua University, Beijing, China, Aug 19–23, 1994.
4. Fillipov, V.E.; Sosnin, L. Yu. Modelling of Gas Flow and Separation Process of Multicomponent Mixture of Isotopes in Countercurrent Centrifuge with Internal Input of Feed. In Proceedings of the Fourth Workshop on

Separation Phenomena in Liquids and Gases; Ying, C., Ed.; Tsinghua University, Beijing, China, Aug 19–23, 1994.
5. Borisevich, V.D.; Levin, E.V.; Yupaatov, S.V.; Aisen, E.M., Numerical Investigation of the Separation of Sulfur Isotopes in a Single Gas Centrifuge. At. Energy, **1994**, *76* (6), 454–458.
6. Ying, C.; Guo, Z. Some Characteristics for Multiicomponent Isotope Separation. In *Proceedings of the Fourth Workshop on Separation Phenomena in Liquids and Gases*; Ying, C., Ed.; Tsinghua University, Beijing, China, Aug 19–23, 1994.
7. Ratz, E.; Coester, E.; deJong, P. Production of Stable Isotopes by Gas Centrifuge. In *Proceedings of the International Symposium on Synthesis and Applications of Isotopes and Isotopically Labeled Compounds*; Toronto, Sept 3–7, 1991.
8. Kai, T. Theoretical Analysis of Ternary UF_6 Gas Isotope Separation by Centrifuge. J. Nucl. Sci. Technol. **1984**, *20* (6), 491–502.
9. Harnik-Snijders, H.; Solving the Diffusion Equation for Multiisotope Mixture. In *Proceedings of the Third Workshop on Separation Phenomena in Liquids and Gases*; Wood, H., Ed.; Charlottesville, Virginia, Aug 16–20, 1992.
10. Levin, E.V.; Ying, Chuntong. The Effect of Multicomponent Isotope Mixture Mole Fractions on the Separative Parameters of a Gas Centrifuge. In *Proceedings of the Fourth Workshop on Separation Phenomena in Liquids and Gases*; Ying, C., Ed.; Tsinghua University, Beijing, China, Aug 19–23, 1994.
11. Levin, E.V.; Ying, Chuntong. Separation of Multicomponent Isotopic Mixtures in a Gas Centrifuge—Approximate Method for Solving the System of Diffusion Transport Equations and Analysis of Some Separation Characteristics. Atomic Energy **1994**, *14* (4), 760–767 (a translation from Russian).
12. Wood, H.G.; Mason, T.C.; Soubbaramayer, Multi-isotope Separation in a Gas Centrifuge Using Onsager's Pancake Model. Separation Science and Technology **1996**, *31* (9), 1185–1213.
13. Ying, C.; Guo, Z.; Wood, H. Solution of the Diffusion Equation in a Gas Centrifuge for Separation of Multicomponent Mixtures. Separation Science and Technology **1996**, *31* (18), 2455–2471.
14. Kobayashi, N.; Tomikkawa, E.; Enokida, Y.; Yamamoto, I.; Kai, T. Numerical Analysis of Concentration Profiles of 4-Componnnent Isotope Mixture within Gas Centrifuges. In *Proceedings of the Sixth Workshop on Separation Phenomena in Liquids and Gases*; Yamamoto, I., Ed.; Nagoya University, Nagoya, Japan, Oct 18–21, 1998.
15. Wood, H.G.; Morton, J.B. Onsager Pancake Approximation for the Fluid Dynamics of a Gas Centrifuge. J. Fluid Mechanics **1980**, *101*, 1–31.

16. Wei, J. Solution to Onsager's Equation and Optimization of Parameters for Gas Centrifuge. Ph.D Dissertation, Tsinghua University, Beijing, China, 1993.
17. Wood, H.G.; Jordan, J.A.; Gunzburger, M.D. The Effects of Curvature on the Flow Field in Rapidly Rotating Gas Centrifuges. J. Fluid Mechanics **1984**, *140*, 373–395.
18. Gunzburger, M.D.; Wood, H.G.; Jordan, J.A., A Finite Method for Gas Centrifuge Flow Problem. SIAM J. Sci. and Stat Comput. **1984**, *3* (1), 78–94.
19. Cohen, K.P. *The Theory of Isotope Separation as Applied to the Large Scale Production of U-235*. McGraw-Hill: New York, 1951.
20. Hoglund, R.L.; Shacter, J.; Von Halle, E. *Kirk-Othmer Encl. Chem. Tech.* 3rd Ed., **1978**, *7*, 695.

Received January 7, 2000
Revised May 2000